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Unidirectional Laser Emission from Spiral-Shaped Microdisk Based on Conducting Polymer

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Introduction

Conducting polymers with highly extended conjugated π -electron systems in the main chains have attracted great interest from both fundamental and practical viewpoints, because they exhibit various novel properties.¹⁾ Among various conducting polymers, poly(*p*-phenylenevinylene) (PPV) and its derivatives are the most attractive materials for light-emitting diodes,^{2,3)} because of their high solubility in common organic solvents and the high luminescence quantum efficiency. Previously, we also reported on spectral narrowing and lasing phenomena in the film of PPV derivatives, poly(2,5-dialkoxy-*p*-phenylene vinylene) (ROPPV), by pulsed photo-pumping.⁴⁻⁶⁾

Polymer lasers with microcylindrical structures have attracted much attention as a novel type of lasers, because of the high Q factors and low threshold energy for the lasing. Previously, we have observed a laser emission based on waveguide mode or whispering gallery (WG) mode from microring and microdisk structures of PPV derivatives.^{7,8)} Furthermore, EL devices with microcavity structures have been fabricated utilizing several kinds of conducting polymers.^{6,8)} The laser emission from micro-cylindrical structures, such as microring and microdisk, however, radiates equally to each direction in plane from the cylindrical microcavity. Therefore, unidirectional laser beam is significant for developing the device application in optical communication. Recently, Chern et al. have successfully observed unidirectional emission in spiral-shaped micropillar lasers with inorganic quantum wells.⁹⁾ The investigation of the spiral-shaped microdisk lasers utilizing luminescent conducting polymer, such as PPV derivatives, should be also important for the realization of a plastic laser diode.

In this study, we report on the fabrication of spiral-shaped microcavities consisting of a PPV derivative and the laser emission properties of the spiral-shaped microcavities of the PPV derivative.

Experimental

One of the PPV derivatives, poly(2-methoxy-5-dodecyloxy-*p*-phenylenevinylene) (MDDOPPV), was used in this study and dissolved in chloroform, which is convenient for spin-coating. The molecular structure of the MDDOPPV is

shown in Fig. 1. The details of the preparation of the MDDOPPV were reported previously.³⁾

Spin-coated films of MDDOPPV, thickness of which were approximately 400 nm, on quartz substrates were etched into spiral shape, defined by

$$r(\theta) = r_0 \left(1 + \frac{\epsilon}{2\pi} \theta \right) \quad (1)$$

by photolithographical etching. Here, ϵ is the deformation parameter. r_0 is the radius of the spiral at $\theta=0$. The angle of $\theta=0$ corresponds to a direction parallel with the notch line, which indicates a line based on the short gap between the radiuses at $\theta=0$ and at $\theta=2\pi$. The notch of spiral-shaped microdisk is significant to obtain unidirectional laser emission. Electron micrographs of the microdisks were taken with a Hitachi S-2100 scanning electron microscope (SEM) to confirm the fabricated spiral-shapes.

For the laser measurement, we used a Nd: yttrium aluminum garnet (YAG) regenerative laser amplifier producing 100 ps pulses with a repetition rate of 1 kHz. This laser light was frequency doubled (532 nm). The pump laser beam was focused by a round lens to be a beam spot with the diameter of approximately 200 μm onto the microdisks of polymer films. The emission from the microdisks was detected by a spectrometer and charge-coupled device array with spectral resolution of approximately 0.2 nm. The samples were held in an evacuated quartz vessel, which could be rotated in the plane of the microdisk.

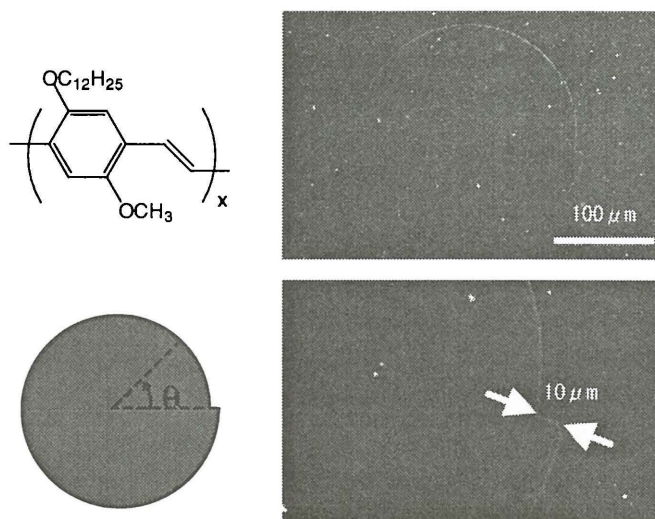


Fig. 1. Molecular structure of MDDOPPV, schematic top-view of spiral-shaped microdisk, and SEM images of a typical spiral-shaped microdisk.

Results and discussion

Figure 1 also shows the schematic top-view of spiral-shaped microdisk and SEM images of a typical spiral-shaped microdisk, which was fabricated by using a photomask with $r_0 = 100 \mu\text{m}$ and $\epsilon = 0.1$ in the photolithographical process. The wall profile, as well as the top surface, shows a smooth face. Such smooth shapes could be also obtained at the notch of the spiral-shaped microdisk, as be obvious in this figure. The notch width in the fabricated microdisk was measured to be approximately 10 μm .

The laser emission from the spiral-shaped microdisk could be observed by the excitation energy higher than the threshold energy. In the case of $\theta=60^\circ$, the threshold energy per pulse was approximately 30 $\mu\text{J}/\text{cm}^2$, and the multi-mode emission, the intensity of which increased linearly above the threshold, could be observed as shown in Fig. 2. It is noted that several sharp laser lines could be observed, but the multimode spectrum was quite different from the typical regular

WG mode from a normal circular microdisks.^{7,8)} The threshold energy changes depending upon the angles in the plane of the disk for the detection. In the case of $\theta=270^\circ$, for example, the threshold energy per pulse was approximately $60 \mu\text{J}/\text{cm}^2$, and the broad photoluminescence spectrum was observed as shown in Fig. 3, because of the excitation energy around the threshold energy. In the case of the excitation energy above the threshold energy, for example, $85 \mu\text{J}/\text{cm}^2$, sharp laser lines could be observed at the similar wavelengths with those of Fig. 2 (a), as shown in Fig. 3 (a). Those emission intensities were much weaker than those in the case of $\theta=60^\circ$.

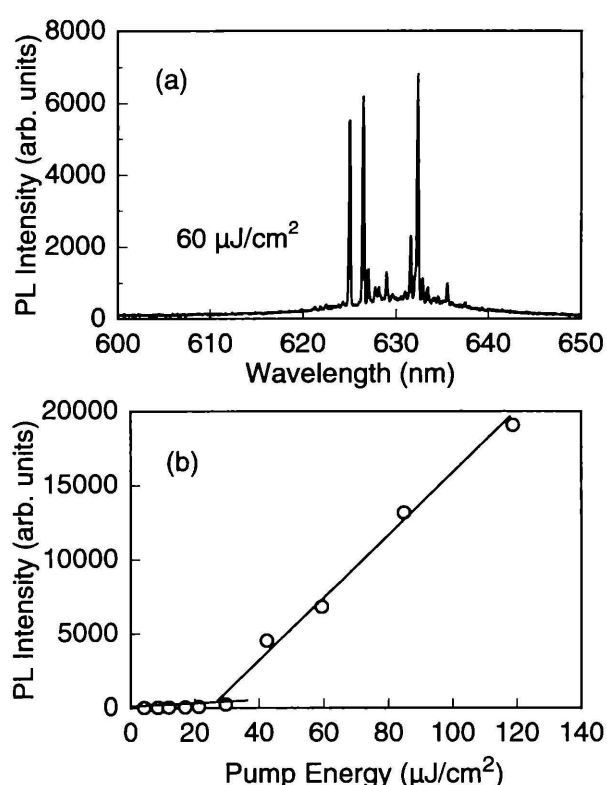


Fig. 2. Emission spectrum (a) and the excitation dependence of the emission intensity (b) in the spiral-shaped microdisk structure of MDDOPPV in the case of $\theta=60^\circ$.

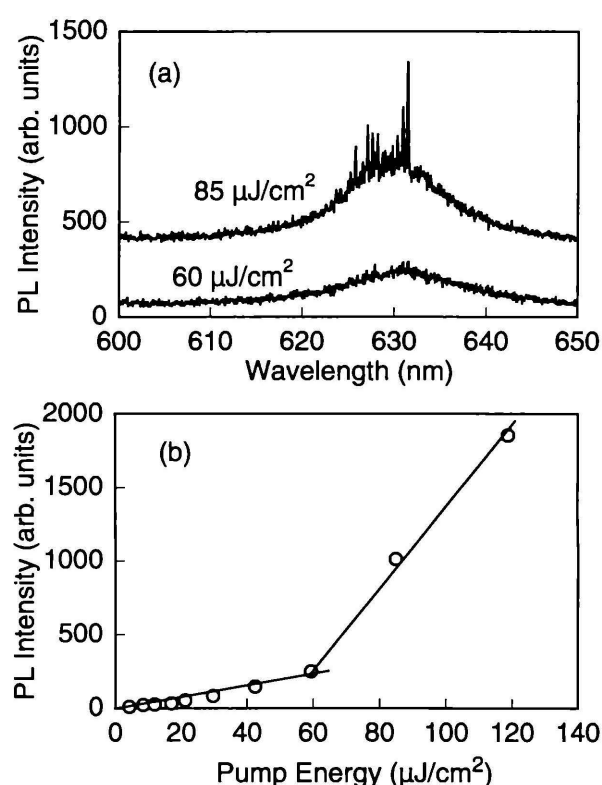


Fig. 3. Emission spectrum (a) and the excitation dependence of the emission intensity (b) in the spiral-shaped microdisk structure of MDDOPPV in the case of $\theta=270^\circ$.

Figure 4 shows the radial distribution of the light output from the spiral-shaped microdisk for various excitation energies. Below the threshold energy for the excitation, the emission pattern was independent of the detecting angle, resulting in the essentially isotropic properties. Above the threshold energy, directional laser emission is clearly observed with the emission direction at a tilt angle between 40° and 70° . It is considered that the tilt in the escape angle of the emission from the notch is due to the spread in wave vectors from the different counterclockwise modes.¹⁰⁾ Though the output beam should escape in a direction which is basically normal to the notch line, these beams with complicated multimodes based on the microcavity with the asymmetric shape might be radiated across the notch

interface with the tilt angles in the range between 40° and 70°.

It should be mentioned that the spiral-shaped microdisk with conducting polymer should be a candidate of future plastic lasers, because it could be fabricated by the simple and quick technique. The simple preparation technique must be effective to fabricate various types of spiral-shaped microcavities easily. The similar type of spiral-shaped microcavity utilizing another luminescent conducting polymer might be also fabricated to obtain blue or green laser emission.

Conclusion

The spiral-shaped microdisks based on MDDOPPV were fabricated on quartz substrates and the laser emission properties of the spiral-shaped microdisk were investigated. The unidirectional red laser emission from the spiral-shaped microcavity composed with MDDOPPV could be observed by pulsed photo-pumping successfully.

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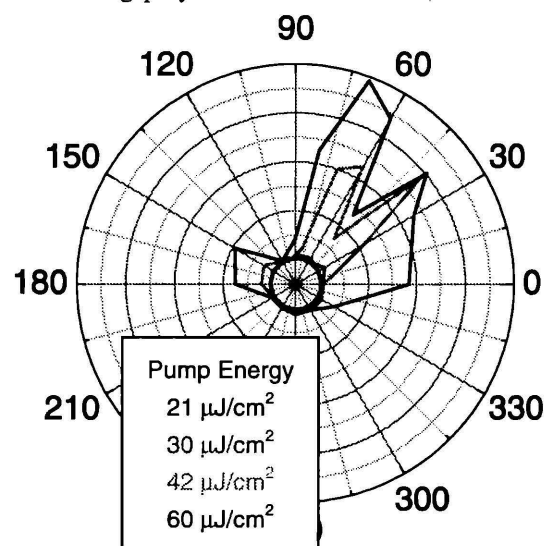


Fig. 4. Radial distribution of the light output from the spiral-shaped microdisk for various excitation energies.